

Energy Correlation in Above-Threshold Nonsequential Double Ionization at 800 nm

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We have investigated the energy correlation of the two electrons from nonsequential double ionization of helium atom in 800 nm laser fields at intensities below the recollision threshold by quantum calculations. The circular arcs structure of the correlated electron momentum spectra reveals a resonant double ionization process in which the two electrons transit from doubly excited states into continuum states by simultaneously absorbing and sharing excess energy in integer units of the photon energy. Coulomb repulsion between the two electrons in continuum states is responsible for the dominant back-to-back electron emission and two intensity-independent cutoffs in the two-electron energy spectra.

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Electron-electron correlation in nonsequential double ionization (NSDI) of atoms and molecules by a short laser pulse at near-infrared (NIR) wavelengths has become the standard example for studies of dynamical electron correlations, which govern the dynamics of many phenomena in nature [1]. Thus, it has been investigated extensively both in experiment [1, 2, 3, 4, 5, 6] and in theory [8, 9, 10, 11] in the past two decades. The physical mechanism responsible for NSDI is well established via the classical "recollision model" [12]. In this model, the first ionized electron is driven back to its parent ion by the oscillating laser field, causing the ionization of the second electron in a direct ($e, 2e$)-like encounter or indirectly via recollision-induced excitation of the ion plus subsequent field ionization (RESI) [4].

At high laser intensities, two recent experiments [13, 14] revealed some novel dynamical details in ($e, 2e$) process. The fingerlike structure in the correlated electron momentum spectrum from NSDI of He by a 800 nm,

4.5×10^{14} W/cm² laser pulse is contributed by binary and recoil recollisions [13]. At a higher intensity, the correlated momentum spectrum exhibits a pronounced V-shaped structure, which is explained as a consequence of Coulomb repulsion and typical ($e, 2e$) kinematics [14]. At intensities below the recollision threshold, a very recent experiment of double ionization of Ar [15] found dominant back-to-back emission of the two electrons along the laser polarized direction, in striking contrast to previous findings at higher intensities. In addition, this experiment confirmed a predicted intensity-independent high-energy cutoff in the two-electron energy spectra [16]. However, the detailed microscopic dynamics in RESI process has remained unknown.

Quantum-mechanical calculations of multiphoton double ionization of He at extreme ultraviolet (XUV) wavelengths [17], as well as visible and ultraviolet (UV) wavelengths [18], demonstrated nonsequential double-electron above-threshold ionization from which the two-electron energy spectrum shows peaks separated by the photon energy. Recent experiments on double ionization of He

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and Ne by strong free-electron laser pulses [19, 20] observed a dominant NSDI process in which the two electrons can absorb two vacuum ultraviolet photons resonantly. Unlike NSDI at XUV and ultraviolet wavelengths in which the two electrons transit directly from the ground state into continuum states, at NIR wavelengths they are ionized from excited states into continuum states after recollision for RESI mechanism and this process is sequential according to Ref. [4]. Does the resonant double ionization also exist in NSDI of atoms at NIR wavelengths?

In this Letter, we investigate the correlated electron momentum and energy spectra from NSDI of helium atoms by ultrashort 800 nm laser pulses at intensities below the recollision threshold by numerically solving the two-electron time-dependent Schrödinger equation. Our calculations excellently reproduce the experimental results in [15], and most importantly, reveal that a resonant double ionization process dominates in RESI mechanism. We draw a new NSDI scenario: firstly doubly excited states are formed by recollision, then the electrons simultaneously absorb an integer units number of photons and share excess energy, transiting into continuum states and followed by a release of Coulomb repulsion energy between the electrons in the continuum states. This NSDI scenario can provide reasonable explanations for the back-to-back emission of the electrons and two intensity-dependent cutoffs in the two-electron kinetic energy spectra predicted in our calculations, as well as observed in the experiment [15].

The experiment [15] has shown that at intensities below the recollision threshold no effect of Coulomb repulsion between the two electrons is found in the direction perpendicular to the laser polarization. Hence, we employ a "one-plus-one"-dimensional model of an helium

atom with soft Coulomb interactions, where the motion of both electrons is restricted to the laser polarization direction [9]. We use the split-operator spectral method [21] to numerically solve the two-electron time-dependent Schrödinger equation (in atomic units)

$$-i\frac{\partial}{\partial t}\Psi(z_1, z_2, t) = H(z_1, z_2, t)\Psi(z_1, z_2, t), \quad (1)$$

where z_1, z_2 are the electron coordinates. $H(z_1, z_2, t)$ is the total Hamiltonian and reads

$$H(z_1, z_2, t) = -\frac{1}{2}\frac{\partial^2}{\partial z_1^2} - \frac{1}{2}\frac{\partial^2}{\partial z_2^2} - \frac{2}{\sqrt{z_1^2 + 1}} - \frac{2}{\sqrt{z_2^2 + 1}} + \frac{1}{\sqrt{(z_1 - z_2)^2 + 1}} + (z_1 + z_2)E(t). \quad (2)$$

$E(t)$ is the electric field of a laser pulse. Following Ref. [9], the two-dimensional space is partitioned into two outer regions: (A) $\{|z_1| < a\}$, or $\{|z_2| < a\}$ and (B) $\{|z_1|, |z_2| \geq a\}$ with $a = 150$ a.u. The final results are insensitive to the choice of a ranging from 100 to 200 a.u. In region A, the wave function is propagated exactly in the presence of combined Coulomb and laser field potentials. In region B, which corresponds to double ionization, all the Coulomb potentials between the particles are neglected and the time evolution of the wave function can be performed simply by multiplications in momentum space. The two regions are smoothly divided by a splitting technique [22]. At the end of the propagation, the wave function in region B yields the two-electron momentum and energy spectra from double ionization.

Our calculations use trapezoidally shaped 800 nm laser pulses with a total duration of 10 optical cycles, switched on and off linearly over 2 optical cycles. A very large grid size of 5000×5000 a.u. with a spatial step of 0.3 a.u. is used, while the time step is 0.1 a.u. The very large grid provides sufficiently dense continuum states [18] to yield highly accurate two-electron momentum and energy spectra. The initiate wave function is the two-electron

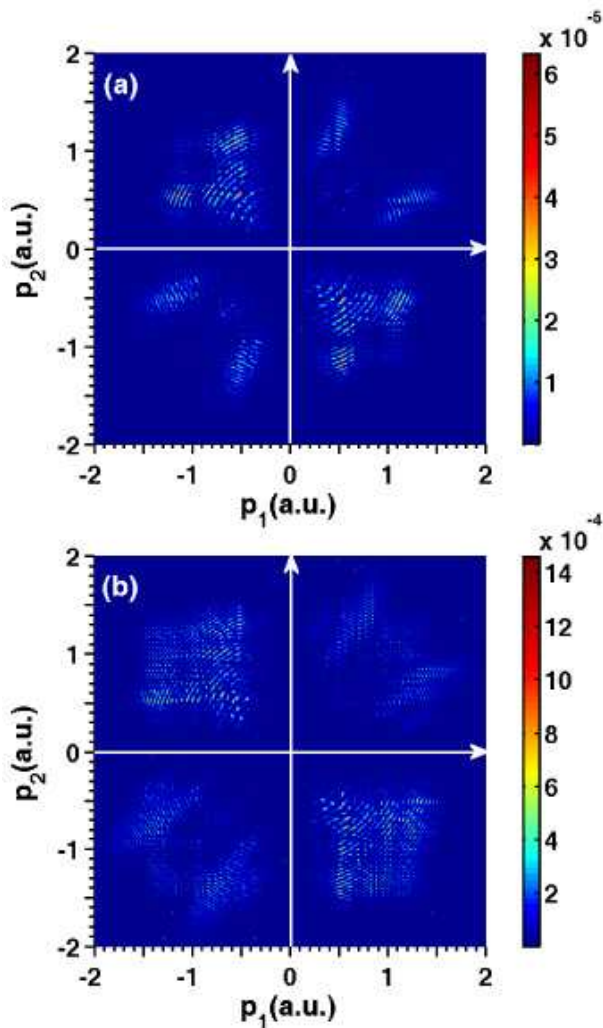


FIG. 1: (color online) Correlated electron momentum distributions for double ionization of He at the intensities (a) 0.1 PW/cm², (b) 0.15 PW/cm². The units are arbitrary.

ground state of He obtained by imaginary-time propagation. After the end of the pulse, the wave function is allowed to propagate without laser field for an additional time of 10 optical cycles. The final results do not change any more even though the wave function propagates without laser field for a longer additional time.

Fig. 1 shows the resulting two-electron momentum spectra from double ionization of He at various intensities below the recollision threshold. These correlated momentum spectra exhibit several significant features. The

first and most striking feature of the momentum spectra is serried concentric arcs, which are most pronounced on a logarithmic scale (see Fig. 2). The concentric arc satisfies $p_1^2 + p_2^2 = \text{constant}$ and it will be shown below that the energy separation between adjacent arcs is the laser photon energy of $\hbar\omega$, where ω is the laser frequency 0.057 a.u. This reveals the dominance of a resonant double ionization process in which the two electrons are strongly correlated. The second feature is that the momentum distributions deviate significantly from the diagonals, which is a sign that the mutual Coulomb repulsion between the two electrons is released in continuum states.

The quantum calculations of double ionization of atoms by strong laser pulses at XUV [17] and UV [18] wavelengths, have shown concentric circles in the correlated electron momentum distributions. However, no effect of Coulomb repulsion is found in the correlated electron momentum distributions. These concentric circles correspond to a resonant double ionization process, in which the strongly correlated two electrons simultaneously absorb and share energy in integer units of the photon energy, transiting from the two-electron ground state into continuum states with the assistance of the Coulomb potential between the two electrons. This process has been called non-sequential double-electron above-threshold ionization (NS-DATI) [17]. Likewise, the first feature found in Fig. 1 also reveals a similar resonant double ionization process. The distinction between the two resonant double ionization processes is that the two electrons transit from the ground state for XUV and UV wavelengths, while from the doubly excited states for NIR wavelength, into continuum states.

The last two features of the momentum spectra in Fig. 1, which have been observed in the experiment [15], are the dominance of back-to-back emission of the two elec-

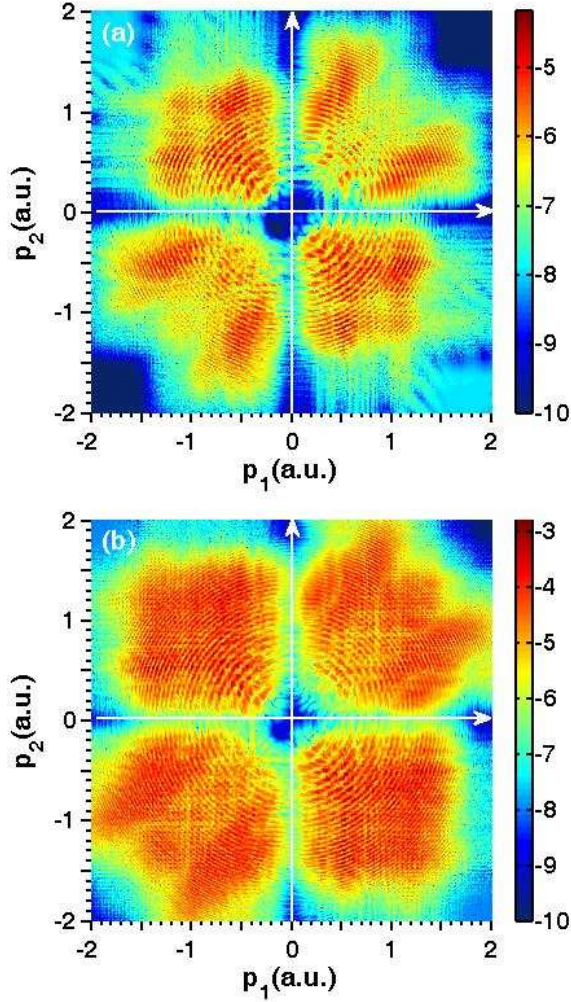


FIG. 2: (color online) Log plot of the correlated electron momentum distributions for double ionization of He at the intensities (a) 0.1 PW/cm², (b) 0.15 PW/cm². The units are arbitrary.

trons) and a clear minimum in a significant area around the origin. The back-to-back emission is considered to be the result of multiple inelastic field-assisted recollisions by the authors of [15]. However, this explanation seems to be unreasonable since the probability of multiple recollisions is extremely low. Here, we consider the release of the strong Coulomb repulsion between the two electrons in continuum states to be responsible for the last three features. More details of our explanation are

given in the proposed scenario below.

According to these dominant features of the correlated electron momentum spectra in Fig. 1 remarked above, we draw a scenario for the corresponding double ionization process. At intensities below recollision threshold since the kinetic energy of the recolliding electron is not enough to directly free the second one, strongly correlated, doubly excited states are formed by recollision. Then, the two electrons transit rapidly from excited states into continuum states by simultaneously absorbing a number of NIR photons and sharing excess energy in units of the photon energy. The Coulomb repulsion between the two electrons can not be released in time in such a rapid process. As a consequence, the effect of Coulomb repulsion takes place along the laser polarization direction in continuum states. Near the field maxima double ionization is most probable and the electrons acquire small drift momenta from the laser field, thus Coulomb repulsion between the electrons leads to the dominance of back-to-back emission and the small drift momenta acquiring from the laser field lead to the electron momentum distributions in the second and forth quadrants deviating from the minor diagonal. When freed near the zero field, the electrons can acquire the same large drift momenta from the laser field. Similarly, Coulomb repulsion between the electrons leads to the electron momentum distributions in the first and third quadrants off the main diagonal. Moreover, Coulomb repulsion between the electrons also results in the minimum at the origin in the momentum distributions.

The double ionization process based on this scenario can be called recollision-induced doubly excitation with subsequent above-threshold double ionization. Within this double ionization scenario, the high-energy cutoffs in the two-electron kinetic energy spectrum can be well

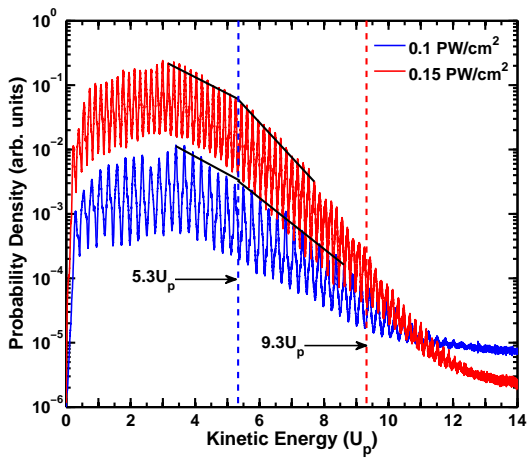


FIG. 3: Two-electron kinetic energy spectra for double ionization of He from Fig. 1.

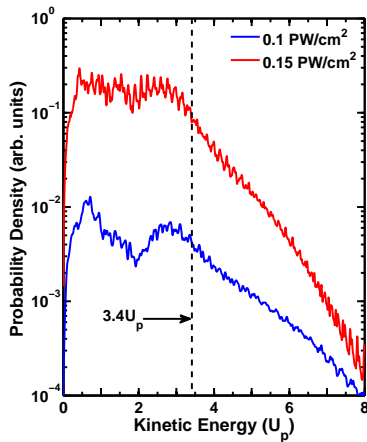


FIG. 4: (color online) Kinetic energy spectra of one ionized electron from double ionization of He from Fig. 1.

explained. Fig. 3 shows the total kinetic energy spectra of both emitted electrons from Fig. 1. Very similar to the above-threshold ionization electron spectrum, the two-electron spectrum also exhibits peaks spaced by the photon energy $\hbar\omega$, implying a strong energy correlation of the two electrons. Moreover, a plateau with an intensity-independent cutoff energy of about $5.3U_p$ is evident in the spectra. Beyond this plateau the spectra decay exponentially until another intensity-independent cutoff energy of about $9.3U_p$. Beyond $9.3U_p$, the multi-

photon effect vanishes quickly and a background plateau follows.

The $5.3U_p$ cutoff has been predicted at 390 nm [16] and observed at 800 nm (see Fig. 1(c) in [15]). The $9.4U_p$ cutoff is also significant in Fig. 1(c) in [15], but not pointed out explicitly by the authors. The difference between the two cutoffs is $4U_p$, just twice of the maximum energy a "free" electron can gain from an oscillating field. Surprisingly, the spectra ranging from $5.3U_p$ to $9.3U_p$ decay exponentially, very similar to single ionization spectra ranging from 0 to $2U_p$ [12, 23]. Thus, we propose that the maximum Coulomb repulsion energy between the electrons corresponds to the $5.3U_p$ cutoff. An additional energy of $4U_p$, the maximum energy the two electrons can gain from the oscillating field, plus the maximum Coulomb repulsion energy, is responsible for the $9.3U_p$ cutoff. Fig. 4 shows an evident intensity-independent cutoff energy of about $3.4U_p$ in the spectra of one electron from double ionization. This reveals that for the first plateau region one electron can share a maximum energy of $3.4U_p$, while the maximum energy of the other can share is $1.9U_p$.

In summary, we have found a new resonant double ionization process at 800 nm and intensities below the recollision threshold, in which both electrons simultaneously absorb and share energy in integer units of the photon energy, transiting from doubly excited states into continuum states. The effect of Coulomb repulsion between the two electrons plays an important role in the electron dynamics in continuum states. This double ionization scenario provides a reasonable explanation for the dominant features in the correlated electron momentum spectra and the two intensity-independent cutoffs in the two-electron kinetic energy spectra. The peaks structure and the two cutoffs in the energy spectra are signatures

of strong energy correlation of both electrons.

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- [1] Th. Weber *et al.*, Nature **405**, 658 (2000).
 - [2] B. Walker *et al.*, Phys. Rev. Lett. **73**, 1227 (1994).
 - [3] Th. Weber *et al.*, Phys. Rev. Lett. **84**, 443 (2000).
 - [4] B. Feuerstein *et al.*, Phys. Rev. Lett. **87**, 043003 (2001).
 - [5] M. Weckenbrock *et al.*, Phys. Rev. Lett. **91**, 123004 (2003).
 - [6] M. Weckenbrock *et al.*, Phys. Rev. Lett. **92**, 213002 (2004).
 - [7] D. Dundas, K. T. Taylor, J. S. Parker, and E. S. Smyth, J. Phys. B **32**, L231 (1999).
 - [8] R. Kopold, W. Becker, H. Rottke, and W. Sandner, Phys. Rev. Lett. **85**, 3781 (2000).
 - [9] M. Lein, E. K. U. Gross, and V. Engel, Phys. Rev. Lett. **85**, 4707 (2000).
 - [10] R. Panfili, S. L. Haan, and J. H. Eberly, Phys. Rev. Lett. **89**, 113001 (2002).
 - [11] C. Figueira de Morisson Faria, H. Schomerus, X. Liu, and W. Becker, Phys. Rev. A **69**, 043405 (2004).
 - [12] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
 - [13] A. Staudte *et al.*, Phys. Rev. Lett. **99**, 263002 (2007).
 - [14] A. Rudenko *et al.*, Phys. Rev. Lett. **99**, 263003 (2007).
 - [15] Y. Liu *et al.*, Phys. Rev. Lett. **101**, 053001 (2008).
 - [16] J. S. Parker *et al.*, Phys. Rev. Lett. **96**, 133001 (2006).
 - [17] J. S. Parker, L. R. Moore, K. J. Meharg, D. Dundas, and K. T. Taylor, J. Phys. B **34**, L69 (2001).
 - [18] M. Lein, E. K. U. Gross, and V. Engel, Phys. Rev. A **64**, 023406 (2001).
 - [19] R. Moshhammer *et al.*, Phys. Rev. Lett. **98**, 203001 (2007).
 - [20] A. Rudenko *et al.*, Phys. Rev. Lett. **101**, 073003 (2008).
 - [21] M. D. Feit, J. A. Fleck, Jr., and A. Steiger, J. Comput. Phys. **47**, 412 (1982).
 - [22] X. M. Tong, K. Hino, and N. Toshima, Phys. Rev. A **77**, 031405 (2006).
 - [23] B. Walker, B. Sheehy, K. C. Kulander, and L. F. DiMauro, Phys. Rev. Lett. **77**, 5031 (1996).